Multiband superconductivity in $PrPt_4Ge_{12}$ single crystals

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We report measurements of the London penetration depth $\Delta\lambda(T)$ and the electronic specific heat $C_e(T)$ on high-quality single crystals of the filled-skutterudite superconductor $PrPt_4Ge_{12}$ ($T_c \simeq$8K). Both quantities show a weak temperature dependence at $T \ll T_c$, following $\Delta\lambda \sim T^n$ ($n \simeq 3.2$) and $C_e/T \sim T^{2.8}$. Such temperature dependences deviate from both conventional s-wave type and nodal superconductivity. A detailed analysis indicates that the superfluid density $\rho_s(T)$, derived from the penetration depth, as well as the electronic specific heat can be consistently described in terms of a two-gap model, providing strong evidence of multiband superconductivity for $PrPt_4Ge_{12}$.

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The filled-skutterudite compounds MT_4X_{12} (M=rare-earth or alkaline-earth metals, T=Fe, Ru, Os, and X=P, As, Sb) demonstrate remarkably rich physical properties [1]. Particular attention has been paid to superconductivity (SC) observed in the Pr-based compounds. For example, $PrOs_4Sb_{12}$ is a heavy-fermion superconductor with T_c = 1.85 K [2]. Electrical-quadrupole, rather than magnetic-dipole, fluctuations are believed to mediate the Cooper pairs in this compound, which is unique among heavy fermion superconductors. The superconducting order parameter of $PrOs_4Sb_{12}$ remains controversial: nodal SC [3, 4] as well as s-wave SC [5] were proposed. More recent experiments seem to support a scenario of multiband SC [6, 7]. On the other hand, the isostructural compounds $PrRu_4Sb_{12}$ and $PrRu_4As_{12}$ appear to be s-wave superconductors [8, 9].

Recently, a series of new skutterudite superconductors with a germanium-platinum framework, i.e., MPt_4Ge_{12} (M=Sr, Ba, La, Pr), were successfully synthesized [10, 11]. Among all the Pr-filled variants, $PrPt_4Ge_{12}$ shows an unexpectedly high transition temperature of T_c=7.9 K [11]. The Sommerfeld coefficient of $PrPt_4Ge_{12}$ (γ_n = 76 mJ/mol K^2) [12] is comparable to that of $PrRu_4Sb_{12}$ [8] and $PrRu_4As_{12}$ [9], but much smaller than that of $PrOs_4Sb_{12}$ [2]. Furthermore, the crystalline electric field (CEF) splitting of the $J = 2$ Hund's rule multiplet of Pr^{3+} between the ground state and the first excited state is rather different among these Pr-based superconductors, e.g., $\Delta_{\rm CEF}$ = 7 K for $PrOs_4Sb_{12}$ [13] and $\Delta_{\rm CEF}$ =130 K in $PrPt_4Ge_{12}$ [11, 14]. It is, therefore, of great interest to systematically compare the superconducting properties of these materials, which may help to elucidate their pairing mechanisms. Similar to $PrOs_4Sb_{12}$, previous studies on polycrystalline samples of $PrPt_4Ge_{12}$ showed controversial results. Measurements of the specific heat and muon-spin rotation (μSR) suggest the possible existence of point nodes in the superconducting gap [12]; zero-field μSR also provides evidence of time reversal symmetry breaking below T_c [15], similar to what was observed for $PrOs_4Sb_{12}$ [16]. However, ^{73}Ge nuclear quadrupole resonance (NQR) experiments display a pronounced coherence peak in the spin-lattice relaxation rate $1/T_1$ at temperatures just below T_c, suggesting s-wave SC [17]. Very recently, a possible scenario of multiband SC was proposed for $PrPt_4Ge_{12}$, based on the analysis of the critical fields [18] as well as photoemission spectroscopy [19]. However, these experiments were performed on polycrystalline samples at relatively high temperatures, which could not make a clear assertion on the gap symmetry. The reasons underlying such discrepancies of the gap structure in $PrPt_4Ge_{12}$ are not yet clear, and further measurements, in particular those based on high-quality single crystals, are badly needed.

In this Letter, we probe the superconducting gap symmetry of $PrPt_4Ge_{12}$ by measuring the London penetration depth $\Delta\lambda(T)$ and the specific heat $C_p(T)$ of high-quality single crystals. Precise measurements of the penetration depth changes at low temperatures show $\Delta\lambda \sim T^n$ with $n \simeq 3.2$, indicating that $PrPt_4Ge_{12}$ is actually neither a simple BCS nor a nodal superconductor. A detailed analysis of the superfluid density $\rho_s(T)$, converted from $\lambda(T)$, and the electronic specific heat $C_e(T)$ provide strong evidence of two-band SC for $PrPt_4Ge_{12}$.

High-quality single crystals of $PrPt_4Ge_{12}$ were synthesized by using multi-step thermal treatments [20]. Powder X-ray diffraction indicates the presence of a small amount of foreign phases. Energy-dispersive X-ray (EDX) analysis confirms that all the crystals have a stoichiometric composition and the impurity phases, mainly $PtGe_2$ and free Ge, are located at the crystal surfaces [20]. In our measurements, the crystals were mechanically polished to get rid of these surface contaminations. Precise measurements of the resonant frequency shift $\Delta f(T)$ were performed by utilizing a tunnel diode oscillator (TDO) based, self-inductance method at an operating frequency of 7 MHz down to about 0.5K

in a ^{3}He cryostat [21]. The change of the penetration depth is proportional to the resonant frequency shift, i.e., $\Delta\lambda(T) = G\Delta f(T)$, where G is solely determined by the sample and coil geometries [4]. In this context, $\Delta\lambda(T)$ is extrapolated to zero at $T = 0$ by polynomial regression, i.e., $\Delta\lambda(T) = \lambda(T) - \lambda_0$. Here the value of zero-temperature penetration depth, λ_0=114 nm, was adopted from previous μSR experiments [12]. Measurements of the magnetic susceptibility were carried out in a SQUID magnetometer (Quantum Design), and the heat capacity was measured in a ^{3}He cryostat, using a relaxation method.

Figure 1 presents the electrical resistivity $\rho(T)$ for $PrPt_4Ge_{12}$, which shows an S-shape behavior upon cooling down from room temperature, as often observed in d-band materials. A sharp superconducting transition, evidenced from both the electrical resistivity $\rho(T)$ (inset (a)) and the magnetic susceptibility $\chi(T)$ (inset (b)), together with a large resistivity ratio (ρ(300K)/ρ(8K)=19) confirm the high quality of our single crystals. Furthermore, the superconducting transition temperatures T_c, determined from the zero resistivity and the onset of the magnetic susceptibility, are nearly the same ($T_c \simeq$7.8 K), proving good homogeneity of the samples.

The inset of Fig. 2 shows the change of the penetration depth $\Delta\lambda(T)$ up to 9K, which reveals a sharp superconducting transition at $T_c \simeq 8.1$ K. Here G=2.13Å/Hz. It is noted that we have measured the penetration depth for several samples and the data are well reproducible. The values of T_c, derived for different samples by distinct methods, are nearly same too. In the main plot of Fig. 2, we presents $\Delta\lambda(T)$ at low temperatures, together with the fits of various models to the data. Obviously, the standard BCS model can not describe the experimental data. Moreover, the penetration depth $\Delta\lambda(T)$ deviates also from that of nodal SC, for which a linear and quadratic temperature dependence is expected for the case of line and point nodes, respectively. Instead, a power law of $\Delta\lambda \sim T^n$ ($n \simeq 3.2$) presents a reasonable fit to the experimental data. An enhanced power-law exponent n, e.g., a quadratic temperature dependence in d-wave superconductors, may arise from nonlocal effects or impurity scattering [22]. However, such possibilities are excluded for $PrPt_4Ge_{12}$ because both the penetration depth (λ_0 =114 nm) [12] and the mean free path (l = 103 nm) are much larger than the coherence length (ξ_0 = 13.5 nm) [12], implying that the samples are in the clean and local limit. Here we estimate the mean free path from $l = [\frac{\xi_0^{-2} - 1.6\times10^{12}\rho_0\gamma_n T_c}{1.8\times10^{24}(\rho_0\gamma_n T_c)^2}]^{1/2}$ [23], where ρ_0, ξ_0 and γ_n represent the electrical resistivity at T_c=7.8K ($\rho_0 = 3.5\times10^{-6}$ Ωcm), the aforementioned coherence length and the Sommerfeld coefficient at T_c (γ_n =1795 erg cm^{-3}K^{-2}), respectively. On the other hand, multiband effects may also give rise to power-law-like behavior at low temperatures with a large exponent n, which will be further elucidated by the analysis of both the superfluid density and specific heat.

The superfluid density $\rho_s(T)$ provides an important characterization of the superconducting gap symmetry. Fig. 3 shows the temperature dependence of the normalized superfluid density $\rho_s(T)$ for $PrPt_4Ge_{12}$ (circles), which is calculated by $\rho_s = [\lambda_0/\lambda(T)]^2$. For comparison, in Fig.3 we also include the superfluid density from the μSR results determined on polycrystalline samples (diamonds) [12]. Obviously, these two data sets are quite compatible, although the μSR data are more scattered, with a poor resolution when compared with the

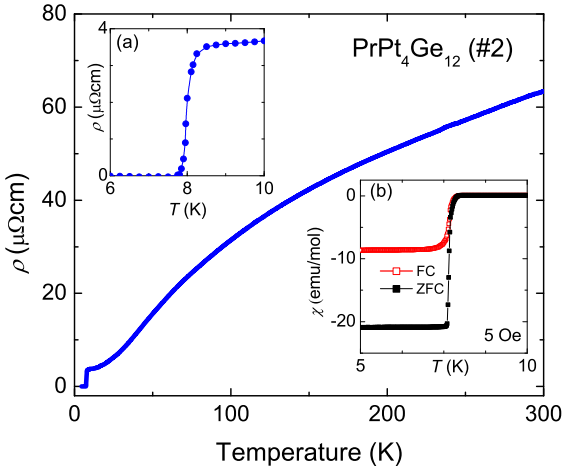

FIG. 1: Temperature dependence of the electrical resistivity $\rho(T)$ for $PrPt_4Ge_{12}$. Insets show the superconducting transitions in the electrical resistivity $\rho(T)$ (a) and magnetic susceptibility $\chi(T)$ (b), respectively.

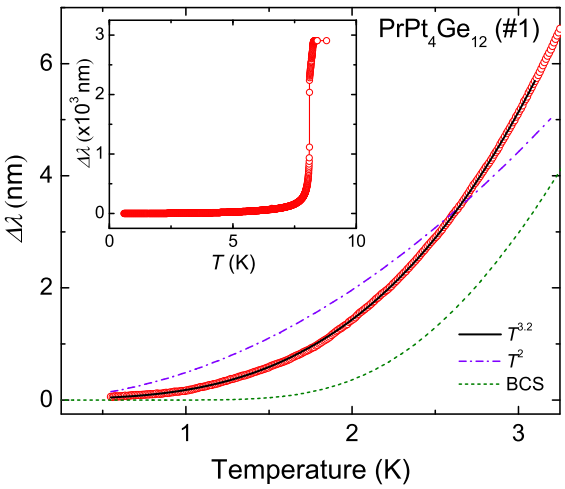

FIG. 2: Temperature dependence of the penetration depth $\Delta\lambda$(T) for $PrPt_4Ge_{12}$. Solid, dash-dotted and dashed lines represent the fit of $\Delta\lambda \sim T^{3.2}$, $\Delta\lambda \sim T^2$ (point node) and single-gap BCS model, respectively. Inset shows $\Delta\lambda(T)$ over the entire temperature range.

TDO results. This allows to probe the gap structure of $PrPt_4Ge_{12}$ in a much more precise way than before.

The superfluid density can be calculated by:

$$\rho_s(T) = 1 + 2\langle \int_0^\infty \frac{\partial f}{\partial E} \frac{E}{\sqrt{E^2 - \Delta_k^2(T)}} dE \rangle_{\rm FS}, \qquad (1)$$

where $f = (e^{\sqrt{E^2+\Delta_k^2(T)}/k_BT} + 1)^{-1}$ is the Fermi distribution function and $\langle \ldots \rangle_{\rm FS}$ denotes the average over the Fermi surface. For the temperature dependence of the energy gap, we take $\Delta(T) = \Delta_0 \tanh \frac{\pi k_B T_c}{\Delta_0} [\frac{2}{3} \frac{\Delta C_e}{\gamma_n T_c} (\frac{T}{T_c} - 1)]^{0.5}$ [24]. Here ΔC_e is the specific heat jump at T_c. Note that Eq. 1 is applicable for various gap functions Δ_k (=$\Delta(\theta, \phi)$) in the pure/local limit. Given a gap function $\Delta(\theta, \phi)$, then one can fit it to the experimental data. Here θ and ϕ denote the angles away from the z-axis and x-axis in k-space, respectively. In this analysis, the zero-temperature gap amplitude, Δ_0, is the sole fitting parameter.

Possible symmetries of the order parameter have been theoretically investigated for the skutterudite superconductors with tetrahedral point group symmetry (T_h) [25]. Various gap functions $\Delta(\theta, \phi)$, restrained by the crystal symmetry, have been adopted to fit the superfluid density $\rho_s(T)$ of $PrOs_4Sb_{12}$ [4]. In this context, we apply a similar analysis to the experimentally obtained $\rho_s(T)$ data of $PrPt_4Ge_{12}$. Fig. 3 presents the fits of different gap functions allowed by the crystal symmetry; the derived fitting parameters of Δ_0 are summarized in Table 1. Apparently, the gap functions B and E cannot reproduce the experimental data. On the other hand, the fits of functions A, C and D are close to the experimental data, but show significant deviations at low temperatures (inset of Fig.3). We note that models C and D, both having point nodes in the superconducting gap, were previously assumed to present a good fit to the μSR data [15], which are rather scattered at low temperature. The more precise measurements of the penetration depth, $\Delta\lambda(T)$, and the corresponding superfluid density, $\rho_s(T)$, indicate that the conventional one-gap BCS model as well as the nodal-gap model D provide a poor fit to the low temperature data. The nodal-gap model C can fit the TDO data relatively well, but significant deviations remain below $0.4T/T_c$. Instead, the two-gap BCS model gives the best fit to our experimental data.

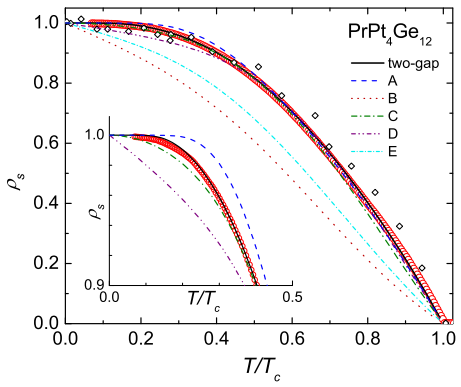

FIG. 3: Superfluid density $\rho_s(T)$ versus normalized temperature T/T_c. Inset expands the low temperature region. Circle (○) and diamond (◇) dispaly the experimental data derived from TDO measurements (this study) and μSR experiments (from Ref.[12]), respectively. Lines shows theoretical fits of various gap functions as listed in Table 1.

TABLE I: Summary of various gap functions and so derived fitting parameters Δ_0.

model	Gap function $\Delta(\theta, \phi)$	Δ_0/k_BT_c
A	Δ_0	1.76
B	$\lvert\Delta_0 \sin\theta \sin\phi\rvert$	4
C	$\lvert\Delta_0 \sin\theta\rvert$	2.7
D	$\Delta_0(1 - \sin^4\theta \cos^4\theta)$	2.9
E	$\Delta_0(1 - (\sin^4\phi + \cos^4\phi)\sin^4\theta)$	3.2

In the case of two-gap BCS superconductors, the superfluid density can be extended to the following linear combination[26]:

$$\widetilde{\rho}_s(T) = x\rho_s(\Delta_0^1, T) + (1-x)\rho_s(\Delta_0^2, T), \qquad (2)$$

where $\Delta_0^i (i = 1, 2)$ represent the size of two isotropic gaps at zero temperature, and x is the relative weight of the contributions from Δ_0^1. As shown in Fig. 3, the two-gap BCS model nicely fits the experimental data of $PrPt_4Ge_{12}$ over the entire temperature region. The so derived parameters of $\Delta_0^1 = 0.8k_BT_c$, $\Delta_0^2 = 2.0k_BT_c$ and x=0.15 meet the theoretical constraints that one gap is larger than the BCS value and the other one is smaller [27], as demonstrated in the prototype two-gap BCS superconductor MgB_2 [26].

In Fig. 4, we present the low-temperature specific heat, $C_p(T)$, of a $PrPt_4Ge_{12}$ single crystal. A sharp superconducting transition is observed at $T_c = 7.7$ K, being close to that determined from other experiments. The specific heat data above T_c can be fitted by a polynomial expansion $C_p(T) = \gamma_n T + \beta T^3$. Here $C_e = \gamma_n T$ and $C_{ph} = \beta T^3$ denote the electronic and phonon contributions, respectively. This yields the Sommerfeld coefficient in the normal state, $\gamma_n = 69$ mJ/mol K^2, and the Debye temperature Θ_D =190 K, which are close to those found in case of polycrystalline samples [12]. For polycrystals [12], a pronounced upturn was previously reported in the low-temperature specific heat $C_e(T)/T$. Similar specific heat anomalies were also observed in some as-grown single crystals. A careful examination showed that such an upturn in $C_e(T)/T$ has to be attributed to a nuclear Schottky anomaly caused by the Pr-containing surface contaminations [20]. Indeed, the specific heat anomaly disappears for the polished single crystal as shown in Fig.4(b), allowing us to accurately analyze its low tem-

perature behavior.

The electronic specific heat of $PrPt_4Ge_{12}$, obtained after subtracting the phonon contributions, is presented in the main part and inset (b) of Fig. 4 as C_e/T verse T, together with the fits of various models. As shown in the inset (b), the data can be well described by a power law, $C_e/T \sim T^{2.8}$. This behavior deviates from the quadratic temperature dependence of $C_e(T)/T$ reported in Ref. [12]. The discrepancy is likely to result from the nuclear Schottky anomaly of the polycrystalline samples discussed before. With the previous data [12], a proper subtraction of this Schottky anomaly is difficult and, therefore, deviations from the true specific-heat behavior become likely at low temperatures. Furthermore, the standard BCS-model is not sufficient to fit the experimental data (Fig. 4(b)), while the two-gap BCS model presents the best fit to the $C_e(T)/T$ data (main figure). According to the phenomenological two-gap BCS model, the heat capacity is taken as the sum of contributions from the two bands, each one following the BCS-type temperature dependence [28]. In the main panel of Fig. 4, we plot the contributions from the two superconducting gaps, $\Delta_0^1 = 0.8k_BT_c$ and $\Delta_0^2 = 2.0k_BT_c$, as well as their sum (solid line). The weight contributed from the first gap, Δ_0^1, is x=0.12. All these fitting parameters are remarkably consistent with those obtained from the superfluid density $\rho_s(T)$, providing strong evidence of two-gap SC for $PrPt_4Ge_{12}$.

Evidence of BCS-like SC, including two-gap type, has been observed in several skutterudite compounds. For example, $PrRu_4Sb_{12}$ [8], $PrRu_4As_{12}$ [9] and their non-$4f$ counterparts [12] are believed to be s-wave superconductors. Recent measurements indicate that $PrOs_4Sb_{12}$ is an extreme two-band superconductor [6, 7]; here energy nodes were assumed to exist in the small gap, and the isotropic large gap dominates the superconducting properties near T_c [7], or when a sufficiently large magnetic field is applied [29]. Two-gap BCS SC was also proposed for both $PrRu_4Sb_{12}$ [7] and $LaOs_4Sb_{12}$ [30], the latter one suggesting that $4f$-electrons are not the origin of multiband SC. In $PrPt_4Ge_{12}$, band structure calculations indicate an only minor contribution of the $4f$-electrons to the density of states at the Fermi energy, suggesting that the $4f$-electrons may not be playing a significant role on SC in this compound either [11]. Indeed, the thermodynamic properties and low-lying CEF scheme of $PrPt_4Ge_{12}$ seem to be rather different from those of the heavy fermion compound $PrOs_4Sb_{12}$, but resemble other skutterudite compounds [8, 9]. Indications of two-gap SC for $PrPt_4Ge_{12}$ were also inferred from recent measurements of the upper and lower critical fields [18] and photoemission spectroscopy [19]. Furthermore, multiband SC is compatible with the observations of a coherence peak in the NQR measurements just below T_c [17]. Such a multi-gap structure seems to be characteristic for the skutterudite superconductors; the small gap, either with or without nodes, is rather subtle and can be easily destroyed by external effects, e.g., a magnetic field, so that the large gap is predominant. Recent μSR measurements performed on polycrystalline samples of $PrPt_4Ge_{12}$ showed evidence of time-reversal symmetry breaking [15]. To confirm it and check the possible existence of nodes in the small gap of $PrPt_4Ge_{12}$, it would be important to repeat these measurements with high-quality single crystals. Detailed calculations of its electronic structure are also highly desirable in order to further elucidate the multiband structure in $PrPt_4Ge_{12}$. Moreover, comparative studies of the Pr-based skutterudites and the non-f electron isostructural compounds, e.g., MPt_4Ge_{12} (M = Sr, Ba and La), are necessary to reveal the potential role of f-electrons on SC.

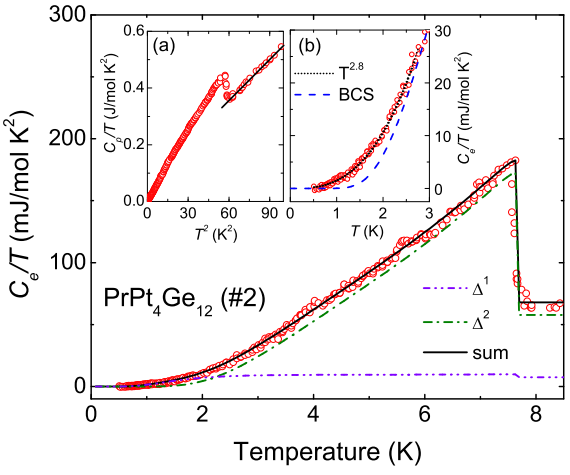

FIG. 4: Temperature dependence of the electronic specific heat $C_e(T)/T$ of a $PrPt_4Ge_{12}$ single crystal at zero field. Dash dotted lines (green and blue) and solid line show the individual and total contributions of the two gaps to the specific heat $C_e(T)/T$, respectively. Inset (a): total specific heat, $C_p(T)/T$, plotted as a function of T^2. Inset (b): electronic specific heat $C_e(T)/T$ at low temperatures, fitted by $C_e/T \sim T^{2.8}$ (dotted line). Dashed line refers to the standard BCS model.

In summary, we have studied the superconducting order parameter of $PrPt_4Ge_{12}$ by measuring the penetration depth $\Delta\lambda(T)$ and specific heat $C_p(T)$ on high-quality single crystals. For $T \ll T_c$, both quantities demonstrate a weak temperature dependence and can be fitted by a power-law behavior with a large exponent, i.e., $\Delta\lambda \sim T^{3.2}$ and $C_e/T \sim T^{2.8}$, which is inconsistent with both a single-gap BCS model and nodal-gap SC. Instead, we can describe the superfluid density $\rho_s(T)$ and the electronic specific heat $C_e(T)$ in terms of a phenomenological two-gap BCS model with consistent gap parameters of $\Delta_0^1 = 0.8k_BT_c$, $\Delta_0^2 = 2.0k_BT_c$ and $x = 0.12 \sim 0.15$, the weight contributed by the small gap. These findings have elucidated the controversial results found in the literature and provide unambiguous evidence of multiband

SC for $PrPt_4Ge_{12}$.

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